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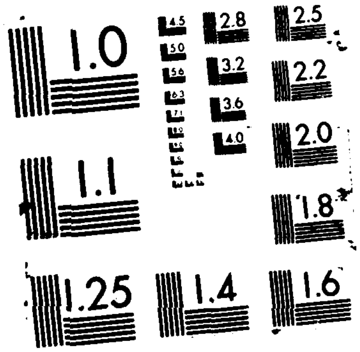
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Doppler Shift Methods for Plasma Diagnostics

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Abstract

Work to develop novel advanced laser spectroscopy plasma diagnostic methods is described. The methods are based on observing the doppler shift in the absorption lines of ionic species. Two methods under study are Velocity Modulated Laser Spectroscopy and Two-Beam Doppler Shift Laser Spectroscopy. The theoretical basis of the methods is described and preliminary experimental results presented.

I) Introduction

The purpose of our work has been to develop advanced laser spectroscopy methods to diagnose partially ionized plasmas. We have focused on methods that are based on observing the Doppler shift in ionic spectra due to the presence of an ion drift velocity. Two particular methods we are working with are Velocity Modulated Laser Spectroscopy (VMLS) and Two Beam Doppler Shift Laser Spectroscopy (TBDSLS).

The scientific goal of our work is to increase understanding of the role of flow non-uniformities and plasma/wall interactions in plasma devices by making in-situ measurements of electric field strength, ion mobilities, concentrations and temperatures in a non-intrusive fashion that allows point, one, and two dimensional imaging.

The scientific approach is to use conventional laser spectroscopic methods such as Rayleigh scattering, Raman scattering, or fluorescence, to probe ion absorption line profiles. If there is an electric field present, the ions will experience a net force and undergo drift, resulting in a shift in the position of the line profile. If the ion mobility is known, then the electric field component along the probe direction can be calculated. If the electric field driving the plasma is modulated, one will observe an oscillating shift in the line profile that arises because of the oscillating force imposed on the ions. The shift may be related to the ion mobility, thus conductivity.

Temperature and concentration may be recovered by conventional laser spectroscopic means. The methods are species and state selective, allowing one to make measurements on more than one species and to study the effect of internal mode nonequilibrium.

The merit of the methods lies in their ability to provide simultaneous measurements of important parameters in plasmas. The methods are well suited to multi-dimensional imaging. One may use an array detector to image lines and planes in addition to the more conventional point configuration.

There has been a far amount of work done by the physical chemistry community on VMLS. VMLS applied to plasmas was

initially exploited by Woods et al who observed Doppler shifts in microwave transitions of HCO^+ [1,2] and then used the shifts to infer ion mobility [3]. Microwave measurement of HCO^+, NO^+, CO^+ and HNN^+ were performed by DeLucia et al [4-7].

VMLS was extended to the infrared region by two groups, that of Professor Richard Saykally of the Chemistry Department at UC Berkeley [8-14] and that of Professor T. Oka of the Chemistry Department at the University of Chicago [15-19]. Saykally's group has focused on the spectroscopy of ions in plasmas while Oka has focused on measurements of ion mobility.

The Chicago group has observed ArH^+, NH_4^+, $HCNH^+$ and $HCOP^+$. The Berkeley group has made observations of H_3O^+, NH_4^+, H_2F^+, HCO^+, HNN^+, and H_3^+.

The Berkeley group has extended the method to the visible region [20-21]. They have observed both CO^+ and N_2^+ in CO or N_2 seeded He plasmas.

The probe method in all the studies so far is absorption spectoscopy. The infrared work was done with a diode laser (Chicago group) and a Krypton pumped color center laser (Berkeley group.) The visible work was done with a single mode dye laser. One of the unique aspects of our work is that we are exploring a number of different probe methods so as to be able to extend the method into the ultraviolet and to molecules that do not absorb at appropriate frequencies.

In the following we describe the theoretical basis of doppler shift methods, outline the nature of the two methods we are pursuing, and describe experimental results to date. The work is then summarized and appropriate conclusions drawn.

II) Theoretical Basis

a) Ion Response to a Modulated Electric Field

The basis of Doppler shift methods is Coulomb's Law which states that the force felt by a charged particle is directly propotional to the imposed electric field

$$\mathbf{F} = q\,\mathbf{E} \tag{1}$$

where E is the electric field, q the charge of the particle and F the force. In a collisional plasma, one may write a momentum equation for the ions that is of the form (in the absence of a magnetic field)

$$d\mathbf{v}/dt = (q/m)\mathbf{E} - \nu_c \mathbf{v} \tag{2}$$

where m is the ion mass, and ν_c the collision frequency. If there is a DC electric field present, then the ion drift velocity will be (after an initial transient)

$$V = \mu_i E \tag{3}$$

where

$$\mu_i = q/m\nu_c$$

is the ion mobility. Likewise, if the electric field is oscillating in form

$$E = E_o \exp(i\omega t) \tag{4}$$

where ω is the angular frequency, , then the velocity will be modulated (after an initial transient) in the form

$$V = (\mu_i/(1+i(\omega/\nu_c)))\ E_o\ \exp(i\omega t) \tag{5}$$

Because of the Doppler effect, the observed resonant absorption or emission frequency of a molecule with a non-zero drift velocity is shifted according to the relation

$$d\nu = (\nu_o/c)\ v \tag{6}$$

where ν_o is the line center frequency, c the speed of light, and v the component of velocity along the line of sight. Thus, for a DC field, the shift will be

$$d\nu/\nu_o = \mu_i E/c \tag{7}$$

and the depth of modulation in resonant frequency due to an oscillating field will be

$$d\nu/\nu_o = ((\mu_i/c)/(1+i(\omega/\nu_c))\ E_o\ \exp(i\omega t) \tag{8}$$

where E and E_o are the component of the electric field along the line of sight in the DC and AC cases respectively. The magnitude of the AC shift will be

$$d\nu/\nu_o = (\mu_i/c)\ (1+(\omega/\nu_c)^2)^{-1/2}\ E_o \tag{9}$$

and the phase of the shift will be

$$\phi = \tan^{-1}(-\omega/\nu_c) \tag{10}$$

Note that in the limit of small ω/ν_c, the modulated result becomes the same as the DC result. When $\omega >> \nu_c$, however, the modulation depth goes to zero and is independent of the collision frequency. Since the experimentalist controls ω, however, this is not a serious problem. If the collision frequency were extremely small the plasma would be in a collisionless domain.

In practice, ions in a hot plasma will have a thermal velocity distribution that will result in a fairly broad absorption line profile. The modulated shift will be easier to see if it is large with respect to the profile. If we take the line width to be given approximately by the Doppler profile we can estimate the ease with which a shift can be seen. The equilibrium Doppler width is

$$d\nu_D/\nu_o = (\sqrt{\ln 2}/c)\ v_t \tag{11}$$

where v_t is the mean thermal velocity. Since the ion collision frequency may be written

$$\nu_c = n_T Q v_t \tag{12}$$

where n_T is the total number density and Q the collision cross section, one may write (if $\omega << \nu_c$ or for the DC case)

$$d\nu/d\nu_D = q/((\sqrt{\ln 2})n_T m Q v_t^2)\ E \tag{13}$$

Invoking the perfect gas law and noting that

$$v_t = (8kT/\pi m)^{1/2} \tag{14}$$

where k is Boltzmann's constant and T the ion temperature, we obtain

$$d\nu/d\nu_D = (\pi q/8Q\sqrt{\ln 2})\ (E/P) \tag{15}$$

where P is the gas pressure.

The coefficient ($\pi q/8Q\sqrt{\ln 2}$) is of order unity in typical plasmas for E in volts per meter and pressure in Pascals. Thus one can obtain significant modulation depths or DC shifts at moderate values of E.

b) The Effect of a Magnetic Field

The effect of a steady magnetic field is to cause the conductivity to become a tensor quantity. In the direction of the magnetic field, however, the conductivity is the same as that given above. Thus by a suitable experimental arrangement one may still utilize the above equations. Furthermore, if the collision frequency is large compared to the plasma gyro frequency, then the tensor conductivity reduces to its zero magnetic field scalar limit.

The plasma gyro frequency is

$$\omega_B = qB/m \tag{16}$$

For singly ionized argon, the value of q/m is about 5.32E6 C/kg, so that for a magnetic field of one Tesla, the gyro frequency would be of the order of a megahertz. Under collisional conditions in most plasmas, the collision frequency will be higher than this. If not, the full equations must be used.

c) Spectroscopic Methods

The choice of a spectroscopic method depends on the spectroscopic properties of the ionic species to be probed and on the thermodynamic state of the plasma.

The candidate methods include single and multi-photon laser induced fluorescence, Rayleigh scattering and electronic Raman scattering (both coherent and incoherent.) If molecular ions are of interest, then rotational and vibrational Raman scattering are possible candidates.

At the present time we must divide ionic species into two spectroscopic classes according to whether their lowest lying electronic states can or cannot be directly excited by available laser systems. Hydrogen and argon are two common propelants whose ionic ground states are extremely difficult to access directly. Barium is an element commonly added as easily ionizable seed with an easily accessed ionic ground state.

The therodynamic state of the plasma is important in determining the possible modes of operation. For a plasma that

is fully ionized, one may utilize non-resonant methods such as Rayleigh scattering because the plasma is almost pure ions and electrons. If the degree of ionization is much less than unity or there is more than one ionic specie present, one must be able to distinguish between species, requiring a resonant method such as laser induced flourescence.

d) Signal Behavior for VMLS

Consider the case in which we chose a spectroscopic method (laser induced flourescence or Rayleigh scattering, for example see Figure 1) that probes the Doppler profile. That is for which the signal is proportional to the Doppler line shape function. If the laser source has a line width much narrower than the Doppler width then the signal will be directly proportional to the Doppler line shape function

$$\mathrm{Sig} \propto (2\sqrt{\ln 2}/\Delta\nu_D\sqrt{\pi})\ \exp-((2\sqrt{\ln 2}/\Delta\nu_D)(\nu-\nu_o))^2 \tag{17}$$

The effect of velocity modulation on the Doppler profile is felt as a modulation of ν_o. By differentiating the signal twice, one obtains the result that the signal is most sensitive to changes in ν_o when $\nu_L = \nu_o \pm 0.425\ \Delta\nu_D$ and the fractional sensitivity is

$$d\mathrm{Sig}/\mathrm{Sig} = 2.35\ d\nu_o/\Delta\nu_D \tag{18}$$

Using the expression for the modulation depth previously derived, we may recast this expression into a detectabilty limit for the detection of a given modulation voltage

$$E_{oDet} = 1.08\ (Q/q)\ P\ (d\mathrm{Sig}/\mathrm{Sig})_{Det} \tag{19}$$

If one assumes a moderate one percent for the signal detectability limit, and a typical ten square Angstroms for the collision cross section, then one obtains

$$E_{oDet}(\mathrm{V/m}) = 0.00675\ P(\mathrm{Pa}) \tag{20}$$

At one atmosphere, the detectable electric field is only 6.75 V/cm, a field readily achievable in the laboratory. By utilizing phase sensitive detection, one can achieve significantly lower detectability limits.

d) Signal Behavior for TBDSLS

In principle, any method which probes the Doppler profile with sufficient resolution can be used to determine a DC line shift. If only one probe is used, however, there may be a problem of absolute frequency calibration. By using two beams in opposite directions, an in-situ calibration is achieved.

Consider the arrangement illustrated in Figure 2. Assume that there is a drift velocity in the positive x direction. If the laser frequency is scanned through the line, then the rightward running beam will probe the profile labeled R in figure 2, while the leftward running wave will probe the profile labeled L.

As the laser frequency is scanned through the line (or lines as it were,) from $\nu < \nu_0$ to $\nu > \nu_0$, signal will first appear from the right running beam. As the frequency is increased, the leftward running beam will begin to contribute. If the two signals are independently observed, they will look like that of Figure 2. (By chopping the beams at a rate fast compared to the frequency scan rate, one may use the same imaging optics to detect both signals.) The peak separation is just twice the Doppler shift and the crossing point of the two signals is the unshifted line center.

For the method to be useful one must also simultaneously determine the ion collision frequency. If one assumes to first order that the collision cross section is constant, then

$$\nu_c \propto P/\sqrt{T} \tag{21}$$

Thus, one may obtain relative profiles without knowing the cross section, or absolute profiles if calibration is possible or the cross section is known. The temperature can be obtained by fitting the line shape of one of the lines to the appropriate Voigt or Doppler profile, or by other means when the line profile is insensitive to temperature. The pressure can be measured by conventional means.

The detectability limit for the method is determined by the resolution with which the distance between the two peaks can be resolved. If the precision with which the line shape signal can be measured is Δs, then for Doppler broadening the precision with which the separation in peaks, and thus electric field, can be resolved is

$$\Delta E/E = (1/\sqrt{2 \ln 2})\ (\Delta\nu_D/\Delta\nu_E)\ (\Delta s/s)^{1/2} \tag{22}$$

If we define the detectability limit as that value of E for which a precision of 10% is achieved when the spectroscopic signal is detected with a precision of 1%, then we can write

approximately

$$E_{det}(V/cm) \approx 0.04\ P(Bar)/T(K)^{1/2} \tag{23}$$

(Assuming that $Q \approx 10A^2$, m is the mass of a proton, and the line center is at 5000A.) Thus at a pressure of 0.01 Bar and 5000K, the detectability limit is about 1 V/cm.

It should be noted that least squares fitting the line shape to the appropriate profile as suggested above, would significantly increase the precision with which the line center is detected and thus reduce the detectability limit for the electric field. Also, if a suitable transition is found, saturation spectroscopy [22] could be used to locate the line peak with high precision, although the in situ calibration for the zero shift position would be lost.

e) Effect of Plasma Conditions

Plasmas of interest in propulsion applications span a wide range of state conditions. At the inlet of a MPD thruster, for example, the pressure is quite high and the gas, as yet un-ionized, is collision dominated. As the gas becomes ionized and the resulting plasma is accelerated and expands through the exhaust nozzle, the pressure drops dramatically until collisions become unimportant. The temperature may start out very low, room temperature in the laboratory, but can rise to values as high as fifty or one-hundred thousand degrees Kelvin.

Pressure and temperature changes affect the diagnostic mainly through changes in the collision frequency and in the ratio of modulation depth to the Doppler width. As long as the former is large compared to the gyro frequency and the latter large enough to detect, the diagnostic will operate.

Figure 3 is a parametric plot of the collision frequency, the ratio of modulation to Doppler width, and the mean free path as a function of pressure and temperature. A collision cross section of ten square Angstroms and an electric field of one hundred volts per centimeter were used to calculate the parameters. As can be seen, as the pressure drops, the modulation depth increases at a much faster rate than the collsion frequency decreases. Thus the flow becomes collisionless long before the measurement fails.

Another consideration is pressure or Stark broadening at high electric field strengths . The methods will still work even if pressure or Stark broadening [23] make an important contribution to the line width, although they are best suited to the Doppler dominated and low ion and electron concentration limits. The detectability limit will merely be increased because it is determined by the peak detection resolution. The broader

peak of a pressure or Stark broadend line is harder to resolve.

The Stark effect will cause shifts in the line position due to both the imposed electric field and the field of electrons and other ions. The DC Stark effect can be accounted for in a straightforward fashion, and at some plasma conditions will dominate the shift.

The presence of a Stark shift due to electrons and ions will be more of a limiting factor. Unless one can calibrate out the shift, it will appear as a systematic error. Of course if the line width is dominated by Stark broadening, then the Stark width can be measured and the contribution of the Stark effect to the measured shift calculated.

III) Experimental Apparatus

Our system is illustrated in Figure 4. We have an argon ion laser (Spectra Physics 171-19) pumped ring dye laser (Coherent 699-21). The ring laser is fully stabilized and has a line width of less that one megahertz. The output of the laser is directed through a Fabry-Perot interferometer (Coherent Model 251 Spectrum Analyzer) as a means of measuring the relative wavelength precisely. A small portion of the beam is also split off and sent to a wavemeter (Burleigh Model WA 10) for absolute wavelength calibration.

The beam is then directed into the test section (Figure 5) in either the VMLS or TBDSLS configuration. In either case, the beam (or beams) pass through small holes in the electrodes.

The signal is detected by observing the fluorescence at ninety degrees (Figure 4). The signal is collected with F/9 optics and passed through a 1/4 meter Jarrell-Ash monochromator. A RCA 1P28 photomultiplier is used as the detector.

The burner is a cappilary, diffusion flame burner designed by Krupa, et. al. [24]. It allows a wide range of operating conditions without the normal difficulty of using acetelyne and oxygen as reactants. The burner is mounted in a low pressure vessel and can be operated down to about 20 Torr.

IV) Experimental Results

We are conducting preliminary experiments in a flame environment using barium as a seed material. This method is attractive because we can create a partially ionized plasma without a discharge. Electrodes are easily inserted into the flame, which is located in a vacuum vessel and can be operated over a pressure range from 2 Bar to about 20 Torr. Barium ion is attractive because it has a strong ground state absorption line at about 450 nanometers.

We have obtained data on the linewidth of the barium ion at atmospheric pressure. Figure 5 shows a typical line shape profile. The series of sharp peaks is the Fabry-Perot signal superimposed to provide a measure of the frequency. The peaks are 1.5 GHz apart, thus the line is about 4.5 GHz wide.

V) Summary and Conclusions

We have presented the theoretical basis of two laser spectroscopy methods for plasma diagnosis based on observing the doppler shift in the absorption lines of ions subject to a drift velocity. The methods are Velocity Modulated Laser Spectroscopy and Two-Beam Doppler Shift Laser Spectroscopy.

Calculations indicate that the methods will lead to excellent detectability for measurement of electric field (or ion mobility), species concentration, and temperature. By using fluorescence detection, the methods are adaptable to line and planar imaging.

Preliminary data on the linewidth of the barium ion are reported.

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Velocity Modulated Laser Spectroscopy

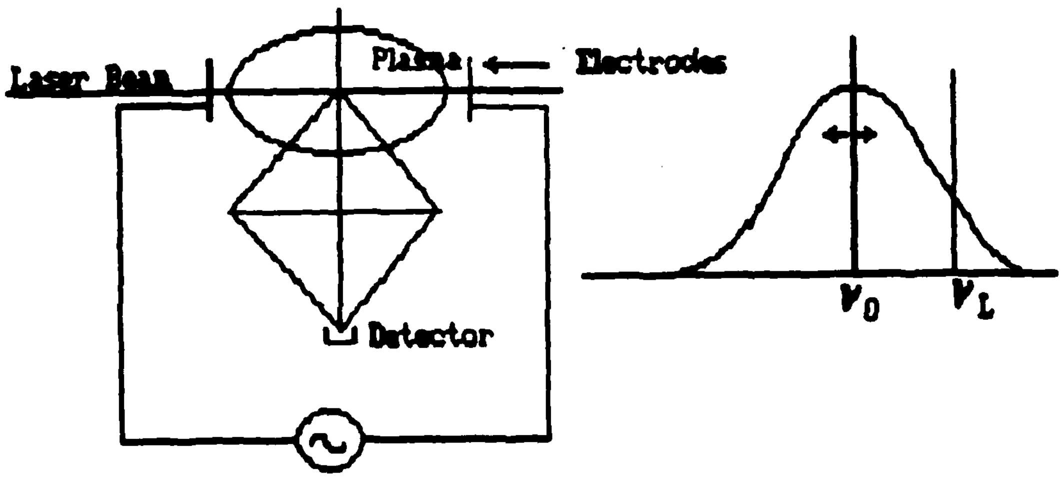


Figure 1.

Two Beam Doppler Shift Laser Spectroscopy

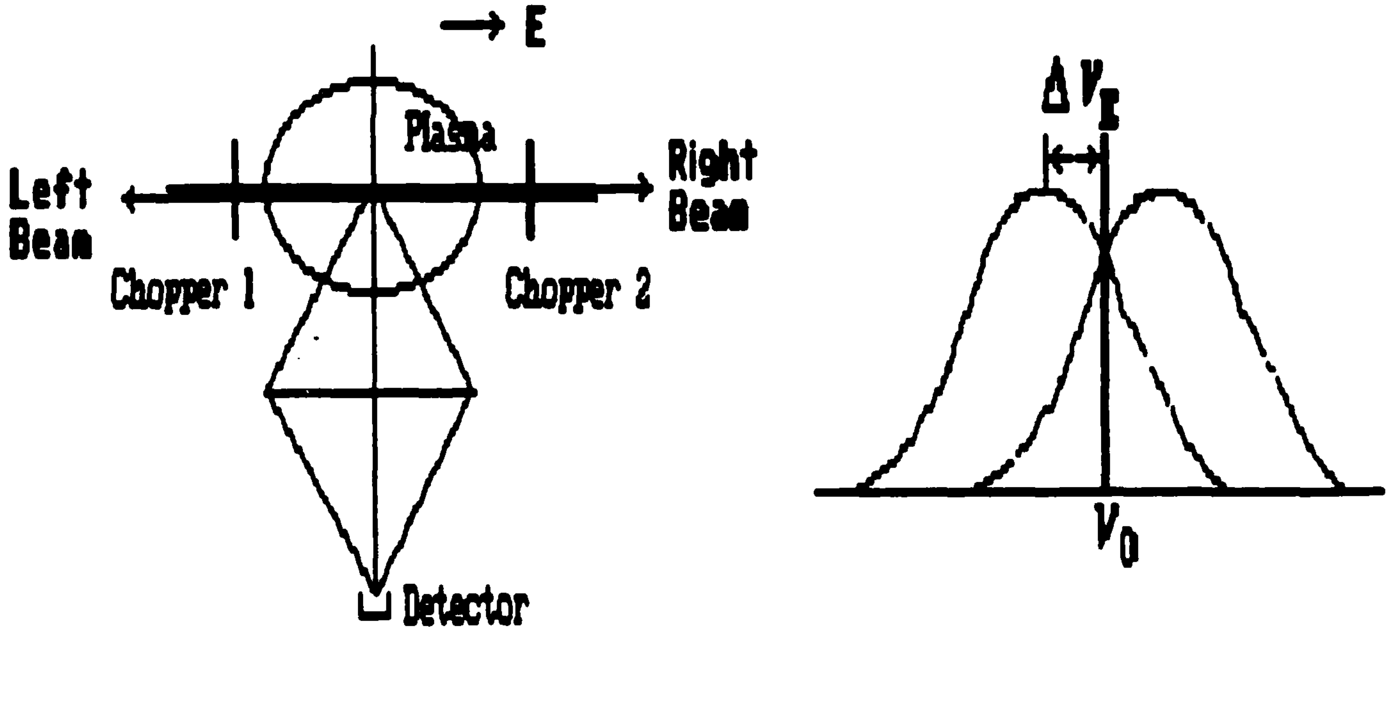
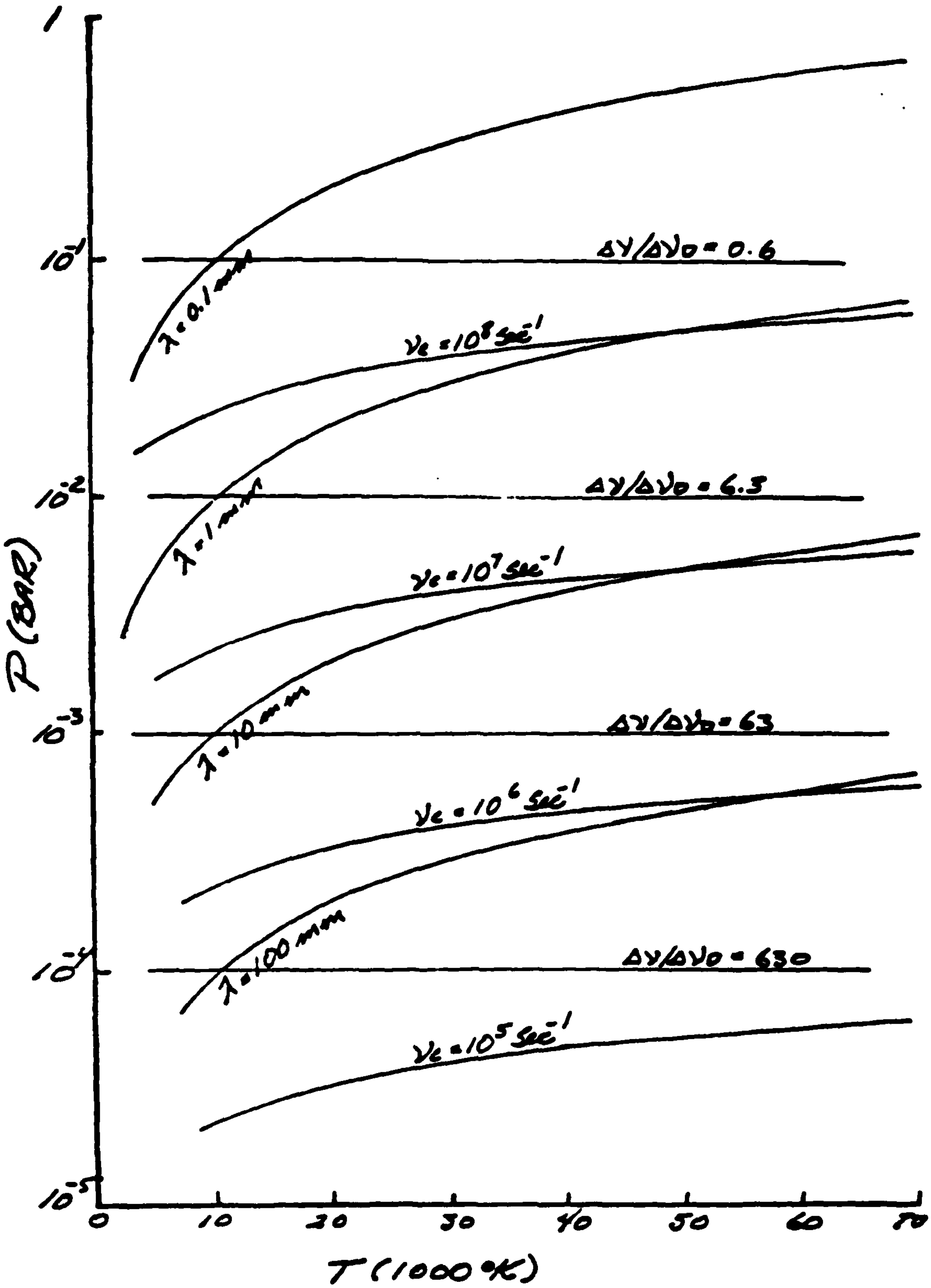
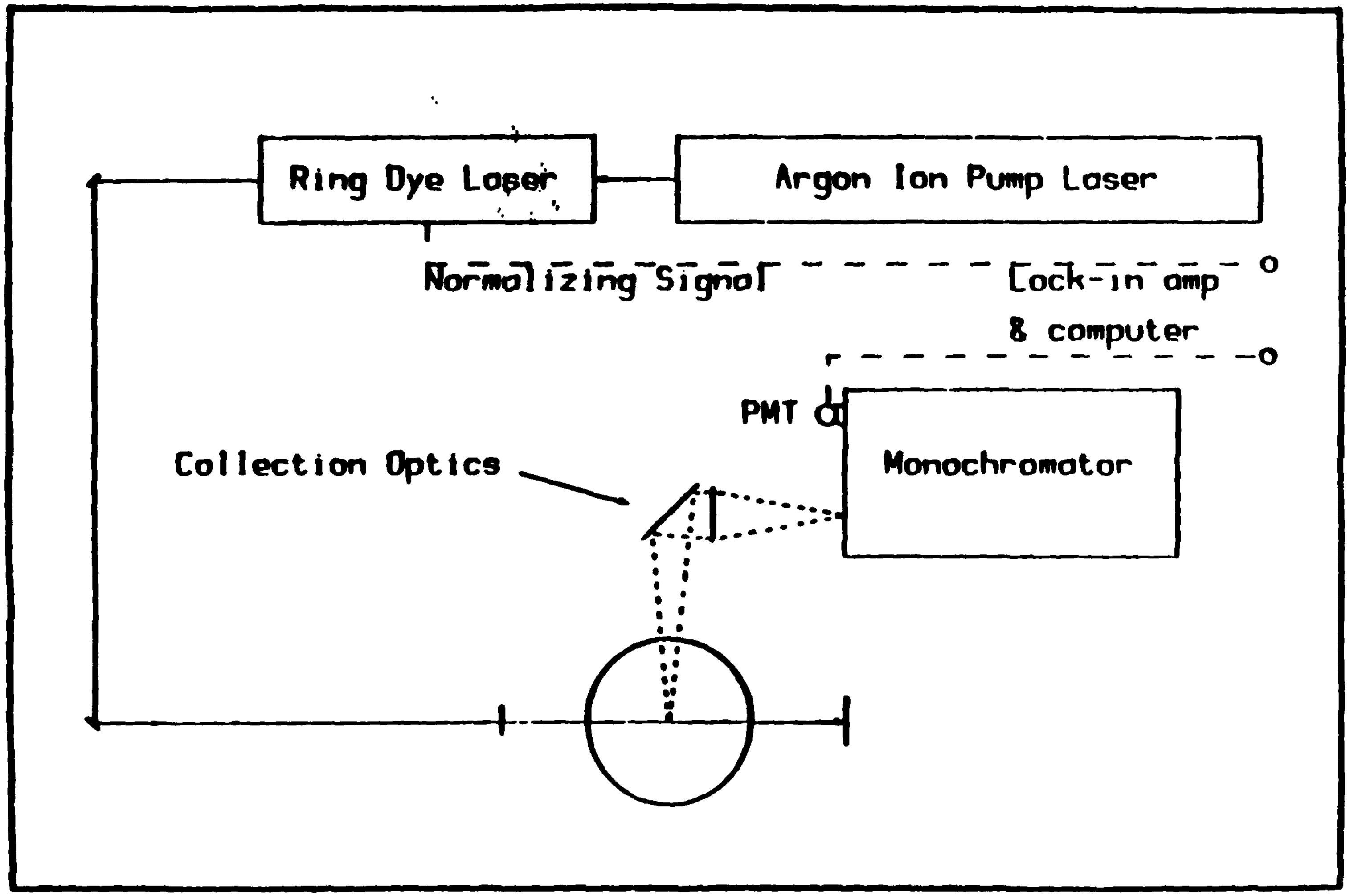


Figure 2.



Parametric Variation of Properties
for VMLS

Figure 3.



Ring Dye Laser
Argon Ion Pump Laser
Normalizing Signal
Lock-in amp
& computer
PMT
Monochromator
Collection Optics

Figure 4.

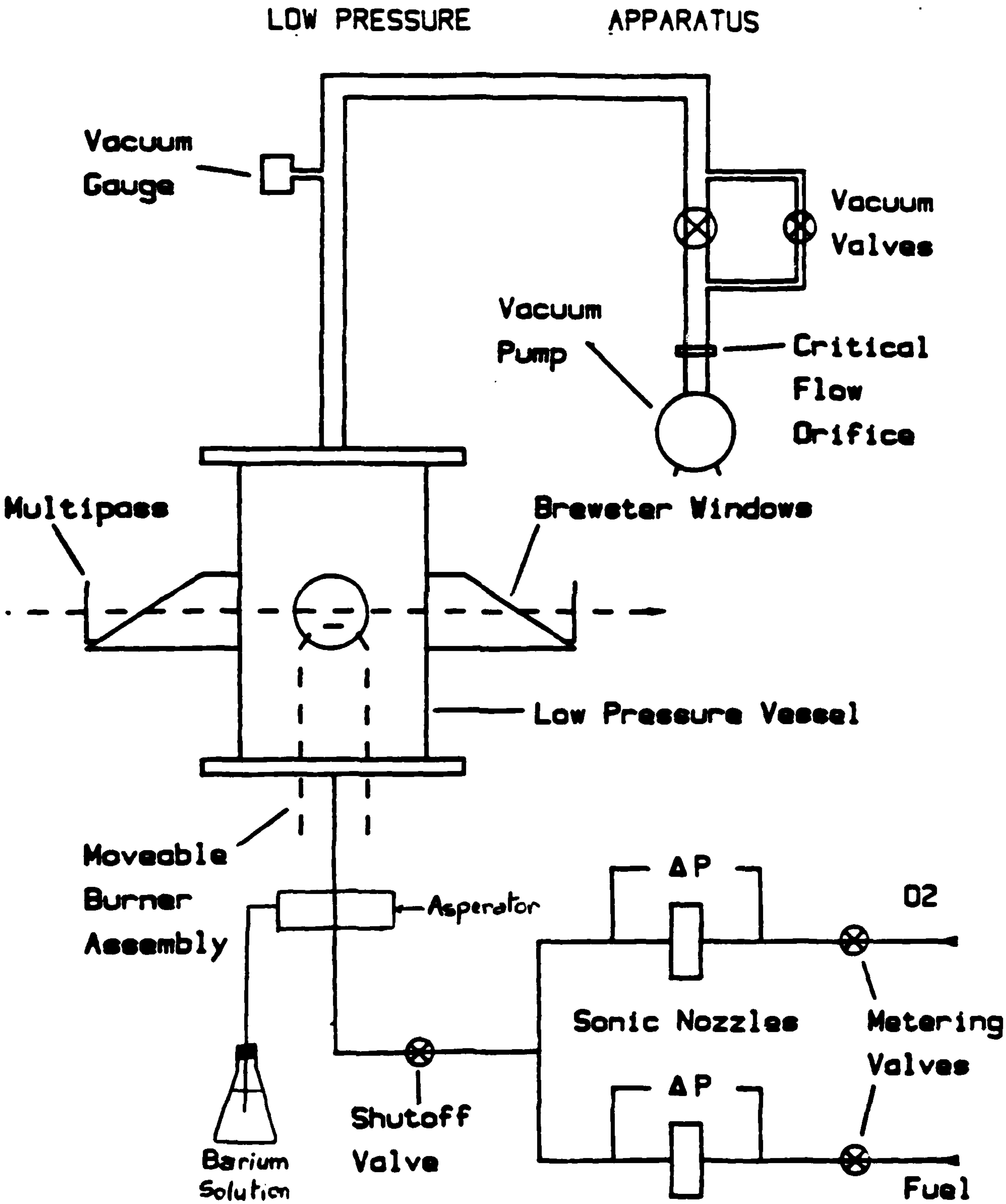
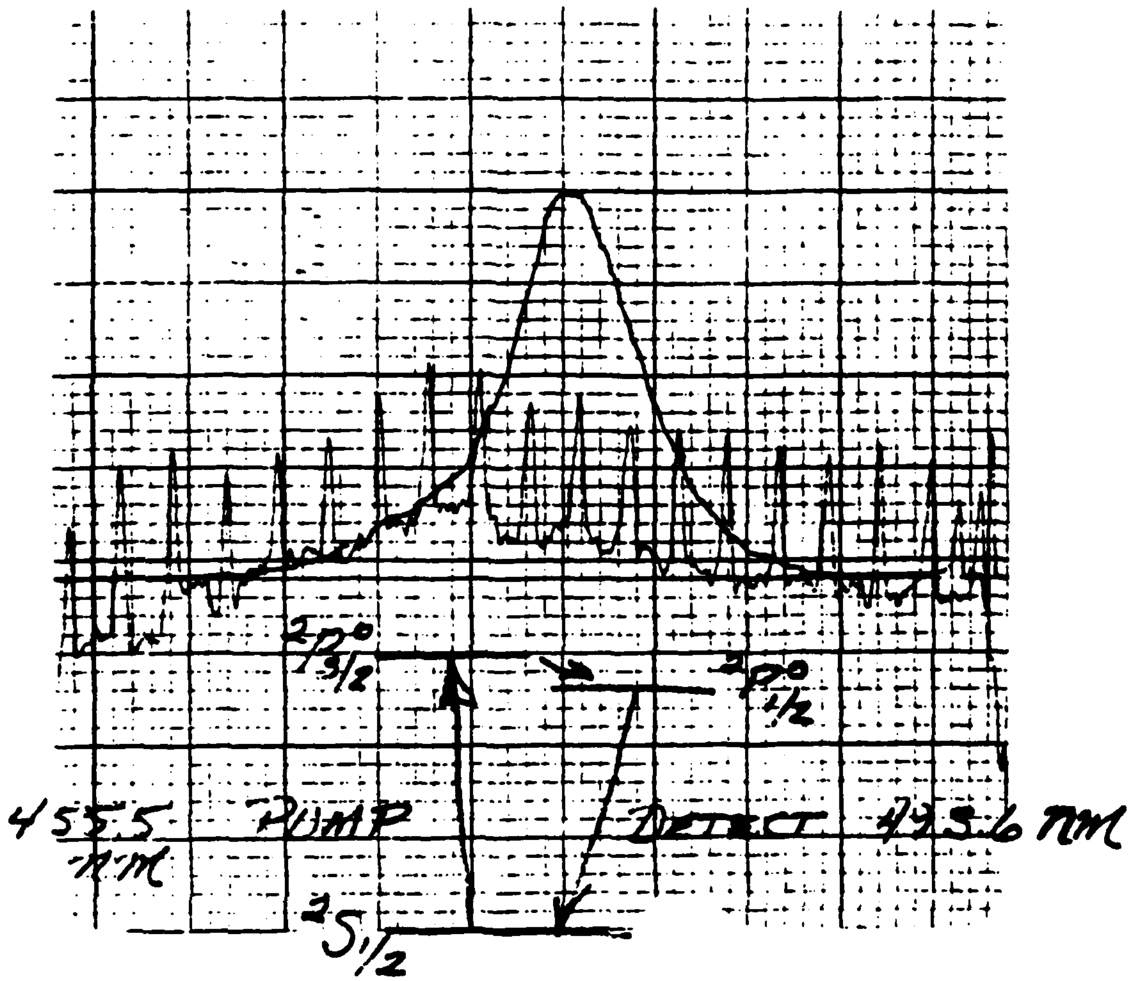

LOW PRESSURE APPARATUS
Vacuum Gauge
Vacuum Valves
Vacuum Pump
Critical Flow Orifice
Multipass
Brewster Windows
Low Pressure Vessel
Moveable Burner Assembly
Asperator
ΔP
O2
Sonic Nozzles
Metering Valves
ΔP
Shutoff Valve
Barium Solution
Fuel

Figure 5.



LIF Signal at atmospheric pressure

Figure 6.

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